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North Pacific-wide spreading of isotopically heavy nitrogen from intensified denitrification during the Bølling/Allerød and post-younger dryas periods: evidence from the Western Pacific

S. J. Kao[1,2], K. K. Liu[2], S. C. Hsu[1], Y. P. Chang[3], and M. H. Dai[4]

[1]Research Center for Environmental Changes, Academia Sinica, Taiwan
[2]Institute of Hydrological Science, National Central University, Jung-Li, Taiwan
[3]Institute of Applied Geosciences, National Taiwan Ocean University, Keelung, Taiwan
[4]State Key Laboratory of Marine Environmental Science, Xiamen University, Xiamen, China

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Correspondence to: S. J. Kao (sjkao@gate.sinica.edu.tw)

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Abstract

Sedimentary $\delta^{15}N$ record for the past 30 ka buried in the Okinawa Trough in the western North Pacific mimicking the pattern from the Eastern Tropical North Pacific (ETNP), but the values (4.4–5.8‰) and the amplitude of the variation were much smaller than those (9–17‰) of the previous site. All but three values in the record were lower than the mean $\delta^{15}N$ (5.6‰) of nitrate in the upper 800 m of the Kuroshio water suggesting additional inputs of isotopically light nitrogen from N_2-fixation. The peak values of $\delta^{15}N$ occurred during the Bølling/Allerød period and the warming period right after the Younger Dryas, synchronous to those found in the Eastern North Pacific. It is highly probable the high $\delta^{15}N$ values are originated from the influence of the intensified denitrification in the ETNP during the warming periods. These new data represent the sedimentary record most distant from the intensive denitrifying zone in the ETNP and may serve as critical constraints to better quantify the nitrogen budget in the last climate cycle.

1 Introduction

As an essential nutrient, changes in the oceanic inventory of biologically available N (or "fixed nitrogen", which is dominated by nitrate) would be expected to impact the biological carbon pump over large regions of the ocean through glacial-interglacial cycles (Falkowski, 1997; Broecker and Henderson, 1998; Archer et al., 2000). Thus, there is a growing interest in better understanding the interaction between climate and N biogeochemistry. Lines of evidence showing strong influences of climate change on the marine N budget have been accumulating (Altabet et al., 1995; Ganeshram et al., 1995; Falkowski, 1997) and many researchers hypothesized indirect influences of the marine N inventory on paleo-climate (McElroy, 1983; Ganeshram et al., 1995, 2000; Pedersen and Bertrand, 2000; Suthhof et al., 2001; Altabet et al., 2002), though considerable uncertainty still remains regarding changes in global ocean nitrate inventory, particularly, during the last glacial period (Deutsch et al., 2004; Altabet, 2007).

N isotopic composition (δ^{15}N) buried in sediments under oligotrophic waters distant from strong denitrification zones may serve as a detector to gauge the magnitude of global denitrification and N inventory changes in the past (Altabet, 2007). One of the unsettled issues on denitrification in the past is its intensity during the last glacial maximum in the North Pacific. Evidence in the South China Sea (SCS), a *cul-de-sac* (Fig. 1a) of North Pacific Intermediate Water (You et al., 2005), supposedly a perfect place to detect the degree of influence from denitrification in the Eastern Tropical North Pacific (ETNP), shows insignificant responses in sedimentary δ^{15}N during the last deglaciation when denitrification peaked in the ETNP (Kienast, 2000; Higginson et al., 2003). To adequately reconstruct past denitrification intensity and changes in marine N inventory more sedimentary δ^{15}N records are needed from oligotrophic regions of the ocean, particularly, the western Pacific.

We present sedimentary δ^{15}N record in the Okinawa Trough. This is the first report showing synchronous δ^{15}N changes in the western North Pacific corresponding to global climate events. Such synchroneity between the western and eastern North Pacific allows us to infer enhanced production of the isotopically heavy nitrogen during intensified denitrification in the ETNP. The amplitude of the isotopic variation may shed new light on the potential changes in the fixed N inventory in the entire North Pacific.

2 Materials and methods

The studied giant piston core, IMAGES-MD012404 (total length 43 m), was recovered at 125.81° E, 26.65° N (Fig. 1b) at a water depth of 1397 m by R/V Marion Dufresne (Bassinot et al., 2002). The coring site is located in a small topographic low near the west edge of the OT, which is ideal for trapping downward settling biogenic particles in the water column, as well as suspended sediments transported from the shelf of the East China Sea (ECS, see Fig. 1b). Sediments in this core are mainly composed of nearly homogenous nanno-fossil ooze or diatom-bearing nanno-fossil ooze and no

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visible turbidite or tephra layer was found in the core (Chang et al., 2005).

The sediment core was sliced into 1 cm thick segments during the cruise and preserved in freezer. A preliminary age model of core MD012404 based on 5 AMS 14C dating for last 30 ka has been published previously (Chang et al., 2005). In this study, a new fine-tuned age model for this core was constructed with additional 14 ^{14}C ages in the last 40000 years (red triangles in Fig. 2a). The AMS ^{14}C measurements were done by taking ~20 mg of the planktic foraminifers *G. ruber* and *G. Sacculifer* (>250 μm), dated at the Micro Analysis Laboratory, Tandem Accelerator (MALT), the University of Tokyo. All AMS ^{14}C ages were adjusted for a mean Pacific reservoir age 400 years, and then calibrated according to Fairbank et al. (2005). No age reversal was observed between any adjacent ^{14}C dating. According to our age model, we also found one interval of carbonate low and ship-board measured magnetic susceptibility high (Bassinot et al., 2002) that coincides with the timing of volcano eruption, Kikai-Ah (~7.3 kyr BP, see Fig. 2a), on the islands of Japan (Machida, 2002).

For $\delta^{13}C_{TOC}$ and $\delta^{15}N$ analyses, 1 N HCl was applied onto the samples for 16 h to remove carbonate; the sediments were then freeze-dried and centrifuged. Details of the sample preservation and pretreatments have been reported in Kao et al. (2006a). The acidified sediments were determined for $\delta^{13}C_{TOC}$ and $\delta^{15}N$ in a Carlo-Erba EA 2100 elemental analyzer connected to a Thermo Finnigan Delta^{plus} Advantage IRMS. Carbon and nitrogen isotopic compositions are presented in the standard δ notation with respect to PDB carbon and atmospheric nitrogen. USGS 40, which has certified $\delta^{13}C$ of –26.24 and $\delta^{15}N$ of –4.52 and Acetanilide (Merck) with $\delta^{13}C$ of –29.76 and $\delta^{15}N$ of –1.52‰, were used as working standards. The reproducibility of carbon and nitrogen isotopic determination for sediment samples is better than 0.15‰. For $\delta^{15}N$, to check if decarbonate process may affect the $\delta^{15}N$ trend we randomly select 11 samples for measuring $\delta^{15}N$ in non-acidified bulk sediment. Result shows consistent values of $\delta^{15}N$ for acidified and non-acidified samples (see Fig. 2b). Contents of total sulfur (TS) reported in Kao et al. (2006a) was taken for discussion.

3 Results and discussion

North Pacific Intermediate Water (NPIW) is a basin-wide distribution (Fig. 1a) of the subtropical salinity minimum (about 34.0–34.3 psu) in a depth range of 300–800 m, confined to the subtropical North Pacific (see You et al. (2005) and references therein). The Okinawa Trough (OT) locates at western Pacific with the Kuroshio Current (KC) entering through the Yonaguni Depression (Fig. 1b) at the southern end of the OT. The bottom depth of Yonaguni Depression ranges from 300 to 800 m, just deep enough to allow the Kuroshio Intermediate Water (KIW) to enter the Trough. The KIW is the main supplier of nutrients to the ECS (Chen, 1996. It has been demonstrated by δ^{15}N values of nitrate that the KIW carries isotopically heavy nitrate originating from the Eastern North Pacific, presumably transported by the NPIW (Liu et al., 1996).

According to recent observations and budget calculation (Chen and Wang, 1999; Liu et al., 2000), the nitrate influx from the KC is 1.5–3.4 times the river load at present. It is conceivable that the primary nutrient source for the ECS in pre-Anthropocene period was dominated by input from the KIW (Liu et al., 2000). Beside the Yonaguni Depression, the opening of Kerama Gap (location shown in Fig. 1b) with bottom depth of 2000 m (though narrow) in the middle Ryukyu Arc is sufficiently deep to afford a part of NPIW exchange.

However, sea-level change in the past (see sea level curve in Fig. 2a, from (Saito et al., 1998 and Liu et al., 2006) may have significant effect on KC volumetric transport, surface and bottom circulations in the OT. Previous studies indicated hydrological conditions significantly altered the sedimentary sulfur and organic carbon biogeochemistry in the OT (Kao et al., 2005; Kao et al., 2006a). TS contents decreased continuously from >0.2% to very low levels (~<0.05%, Fig. 2a) in association with increasing sea levels since 15 cal ka BP. Similar upward decreasing trend in TS content (in terms of age) was also found in core MD012403 from the southern OT (see location in Fig. 1b) despite a ~4 times higher sedimentation rate (Kao et al., 2005). Such synchronous decreases in TS in two cores from the central and southern trough indicate that bottom

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water circulation change is a trough-wide phenomenon. Further down-core diagenetic sulfate reduction might have contributed to the TS profile to some degree; however, the main decreasing pattern is attributed to circulation changes, which have been supported by a 3-D model by (Kao et al., 2006b). Their 3-D model revealed a reduction of KC throughflow (~30% of the throughflow bifurcates before entering the Yonaguni Depression; see Fig. 1b) during the glacial period and KC outlet switches from Tokara Strait to Kerama Gap (see Fig. 1b) leading to weaker deepwater ventilation. However, the Kuroshio Current still enters the Okinawa Trough. Independent evidence shows a transformation from foliation to anomalous sedimentary magnetic fabric (dynamic depositional environment) in the southern Okinawa Trough since the Holocene (Kao et al., 2005). Accordingly, oxygen supply to the deepwater should have been increased as rising of the sea level; by contrast, high TS contents prior to the Holocene may imply higher potential for both water column and sediment denitrification during that period of time.

The $\delta^{13}C_{TOC}$ in MD 012404 ranges from –20.6 to –21.8‰ (not shown) with mean value of –21.2±0.3‰ reflecting a major contribution from marine sources (Goericke and Fry, 1994). A shift in $\delta^{13}C_{TOC}$ was found on the downcore trend between glacial and Holocene periods. Such shift resembles that reported by Kienast et al., (2001) for the northern shelf of the SCS (Core 17940-2, Fig. 1), yet, significantly smaller change in $\delta^{13}C_{TOC}$ (0.5‰ as compared to 1–1.5‰ in Core 17940-2) was observed in the OT. The $\delta^{13}C_{TOC}$ shift trend in the SCS was found basin-widely and unexplained; yet, but changes due to variations in $p\text{CO}_2$ over time or the influx of terrestrial plant organic matter have been ruled out (Kienest et al., 2001). In the Okinawa Trough, the much smaller changes in $\delta^{13}C_{TOC}$ during the same period of time should eliminate the possibility of increased terrestrial inputs, whereas the possibility of intensified upwelling appears to be unlikely on account of such a small variability in $\delta^{13}C_{TOC}$ and the temporal δ^{15}N variation to be discussed next.

The sedimentary δ^{15}N in MD 012404 varies between 4.4 to 5.8‰ (Fig. 2b), which is consistent with the ranges reported for the two cores, 17940-2 and 1144 (see lo-

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cations in Fig. 1b) from the north SCS, respectively, by Kienast (2000) and Higginson et al. (2003). However, the sedimentary $\delta^{15}N$ variations observed in the SCS did not show any recognizable pattern; by contrast, the $\delta^{15}N$ variation found in the Okinawa Trough showed meaningful patterns with good correlation with several other trends, though our study site is rather close to those in the SCS. The low frequency variation of $\delta^{15}N$ in MD 012404 follows the precession cycle in solar insolation at 30^{o}N (Berger and Loutre, 1991) (Fig. 2b) and the $\delta^{15}N$ value of the surface sample agrees well with those values found in surface sediments in the southern Okinawa Trough (Kao et al., 2003). Several high frequency fluctuations with period of ca 2 ka causing notable deviation from the trend of the precision cycle occurred between 8–17 ka BP deserve attention and will be discussed later.

According to Liu et al. (1996), the integrated mean $\delta^{15}N$ of nitrate for the upper 800m of the modern day Kuroshio is 5.6‰ (5.0‰ for upper 400m) and the top 200m is oligotrophic (NO_3 in nM level). If all of the nitrate pumped up to the euphotic zone from the KIW is consumed, the particulate organic nitrogen that sinks back to the seafloor should have the same isotopic composition as the nitrate that gets pumped up, unless there is additional sources of nitrogen, such as nitrogen fixation added to the surface water (Liu et al., 1996). Compared with the integrated mean $\delta^{15}N$ of nitrate in the KIW, all but three sedimentary $\delta^{15}N$ values throughout the entire MD012404 core are lower than the mean $\delta^{15}N$ value of the modern day nitrate reserve in the subsurface water, suggesting that N_2-fixation must have occurred in the Kuroshio surface water most, if not all, of the time over the past 30 ka.

It is noteworthy that the $\delta^{15}N$ variation in MD012404 does not follow the TS trend. As mentioned earlier, the trend of consistently higher TS (higher intensity of sedimentary sulfate reduction) before the last deglaciation suggests hypoxic to anoxic condition in the bottom water of the OT and, therefore, higher potentials of sediment and water column denitrification throughout the pre-Holocene period. Nevertheless, the peak values of $\delta^{15}N$ occurred only in the last few kyr in the pre-Holocene period, but not in the earlier years, when the TS was equally high or even higher, indicating the high sedimentary $\delta^{15}N$ values and denitrification in the bottom water were apparently decoupled. The fluctuation of sedimentary $\delta^{15}N$ values, which reflect the $\delta^{15}N$ of nitrate in the upper water column, must have resulted from influence outside the OT rather than from local sources. Similarly decoupled nitrogen isotopic compositions of nitrate in the upper water column and in the bottom water have also been observed in the sea off southern California (Liu and Kaplan, 1989).

We compare MD012404 record with the sedimentary $\delta^{15}N$ records (Fig. 3a) in the Eastern North Pacific (ENP) during last deglaciation (e.g., JPC-56 at Gluf of California, Pride et al., 1999; ODP-893A at Santa Barbara Basin, Emmer and Thunell, 2000). Record of $\delta^{18}O$ in GISP 2 ice core is plotted (Fig. 3b) for comparison (Grootes et al., 1993). Amazingly, the sedimentary $\delta^{15}N$ mimics the trends observed in the Eastern North Pacific. Despite the much reduced amplitude, the fluctuations of the sedimentary $\delta^{15}N$ in OT followed those in the Gulf of California very closely. It is noted that the $\delta^{15}N$ fluctuations also corresponded to fluctuations in $\delta^{18}O$ in GISP 2 ice core during pre-Holocene period, suggesting close relationship to climate fluctuations. Two $\delta^{15}N$ peaks occur during two warm periods: Bølling/Allerød and the period after Younger Dryas during transgression period (Fig. 3b) resembling those trends observed not only in the ENP but also in the Arabian Sea (Ganeshram et al., 1995, 2000; Deutsch et al., 2004 and references therein), whereas, three $\delta^{15}N$ drops appear during three cold periods, namely, Heinrich Event II (Hemming, 2004), Heinrich Event I and Younger Dryas, either in JPC-56, ODP 893A or in core MD012404. A decreasing trend in $\delta^{15}N$ starts since the Holocene was observed in the OT. Such decreasing trend was reported elsewhere (Higginson et al., 2003; Altabet, 2007; and references therein). Synchronous changes in $\delta^{15}N$ (though much smaller amplitude) in the OT and their close correlation with those warm and cold events and with those temporal patterns in ETNP and Arabian Sea suggest these brief events found in the Okinawa Trough are climate-related and global in nature.

Abundant evidences indicated the oxygen minimum zone (OMZ) in northern and eastern Pacific was more intense during the two warm periods (Behl and Kennett,

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1996; Cannariato and Kennett, 1999; Zheng et al., 2000; Ivanochko and Pederson, 2004; Mckay et al., 2005). Whether the two OMZ intensification events were attributed to increased export production or to suppressed ventilation remain debatable. Recently, a conceptual model regarding O_2 supply to intermediate water and oxygen demand during transportation and their subsequent modulations on denitrification and response of N_2-fixer was provided by Galbraith et al. (2004). The conceptual model explains quite well most temporal variations of δ^{15}N in various oceanic environments.

In our case, synchronous increases in δ^{15}N during the two warm periods indicate the scale of OMZ intensification, and consequently, influence of the enhanced denitrification is likely North Pacific-wide during the last deglaciation. The small amplitude of sedimentary δ^{15}N changes in the OT is possibly due to reservoir dilution (Deutsch et al., 2004) or attenuation caused by N_2-fixation (Deutsch et al., 2007). The only three sedimentary δ^{15}N values in MD012404 exceeding the present day mean δ^{15}N of nitrate in the upper water column of the Kuroshio (Fig. 2b) occurred during the warm periods, when intensification of denitrification in the ETNP. It is conceivable that the nitrate reserve in the upper water column had an elevated mean δ^{15}N value during these warm periods. Therefore, nitrogen fixation should have occurred in the Kuroshio surface water during these warm periods as in other periods over the last 30 ka.

The lack of synchronous changes in sedimentary δ^{15}N records in the northern SCS needs some explanation. It has been reported that N_2-fixation could be important in the SCS (Wong et al., 2002). Basin-wide deep ventilation down to 2000 m occurs in the SCS (Chao et al., 1996). Both processes could have significantly attenuated the denitrification signal from the ETNP. Variations in surface and subsurface circulations and exchanges, such as Kuroshio intrusion, occur due to climate fluctuations (Qu et al., 2004). Similar or more pronounced changes very likely occurred in the past due to climate and sea level changes. Consequently, the inflows of water masses at different density levels, which probably had nitrate with different δ^{15}N values, probably varied considerably in the past. These complicated processes may have led to the unrecognizable patterns of temporal variations of δ^{15}N during the last deglaciation (Higginson et al., 2003). By comparison, the relatively simple gyre circulation in the open North Pacific Ocean resulted in the nice pattern of the δ^{15}N record in the OT.

We may conclude that such substantial δ^{15}N changes in a region very distant from the intensive denitrifying zones in the ETNP lends support to the notion of significant changes in N inventory in the North Pacific in the last climate cycle (Deutsch et al., 2004; Altabet, 2007). However, we cannot rule out the possibility of strong feedback from nitrogen fixation to make up for the nitrogen loss during denitrification. The sedimentary record from our study site, which represents the farthest site, up to now, preserving recognizable signal of δ^{15}N fluctuation from the ETNP, can certainly serve as a valuable constraint for model simulations of the nitrogen cycle during the last glaciation-deglaciation cycle.

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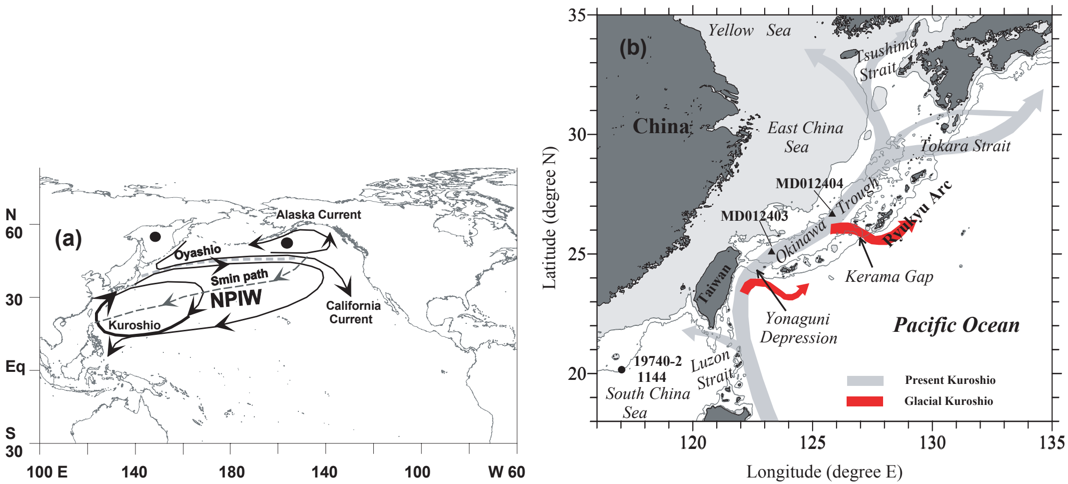

Fig. 1. (a) Geographic setting for the North Pacific. Circulations, path of salinity minimum (S_{min}, see You et al., 2006) and two sources (black circles) of intermediate water are indicated. **(b)** Location map for IMAGES core MD012404 and MD012403. Core 17940 and 1144 in previous reports are also shown. The land (deep gray), shelf of <–100 m (light gray) and –1000 m isobaths are shown. The flow path of present Kuroshio and glacial Kuroshio are shown in gray and red arrows.

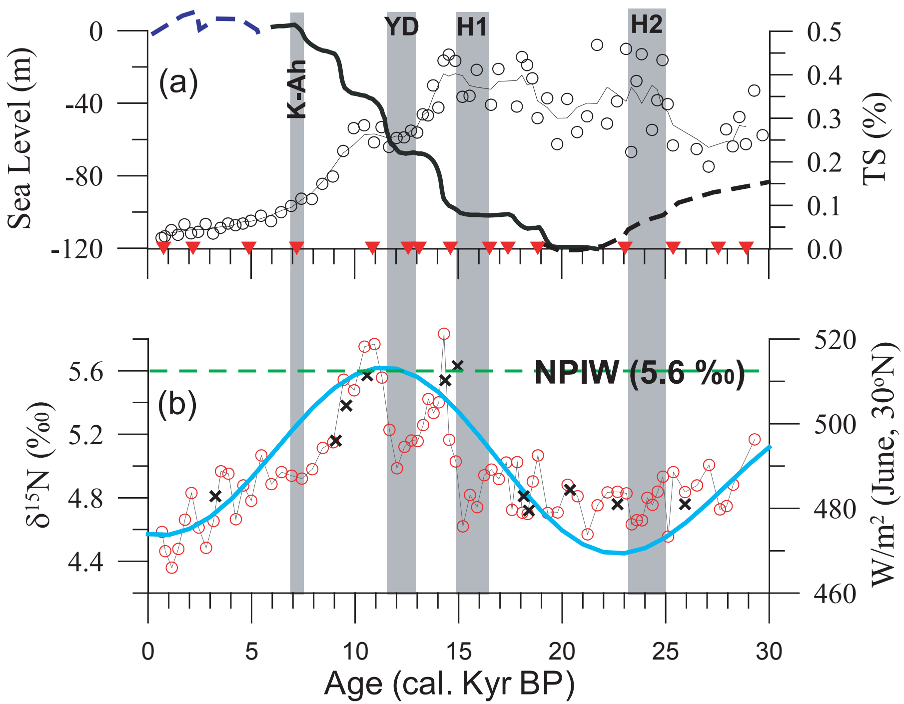

Fig. 2. **(a)** Sea level curve. Blue dotted-line and black curve represent data digitized, respectively, from Saito et al. (1998) and Liu et al. (2006). Dates (▽)and total sulfur content (TS, ◯) are plotted against age. The 5-point running average for TS is shown in curve. **(b)** Temporal variations of sedimentary δ^{15}N (◯) and solar insolation in June at 30°N (indigo blue). Crosses represent δ^{15}N values for selected non-acidified samples. Gray columns stand for time periods of K-Ah volcanic event (K-Ah), Younger Dryas (YD), Heinrich I Event (H1) and Heinrich II Event (H2). Horizontal green dashed line in (b) marks the integrated mean δ^{15}N of NO_3 (5.6‰) in upper 800 m water column for the Kuroshio Current, which is sourced from North Pacific Intermediate Water (NPIW).

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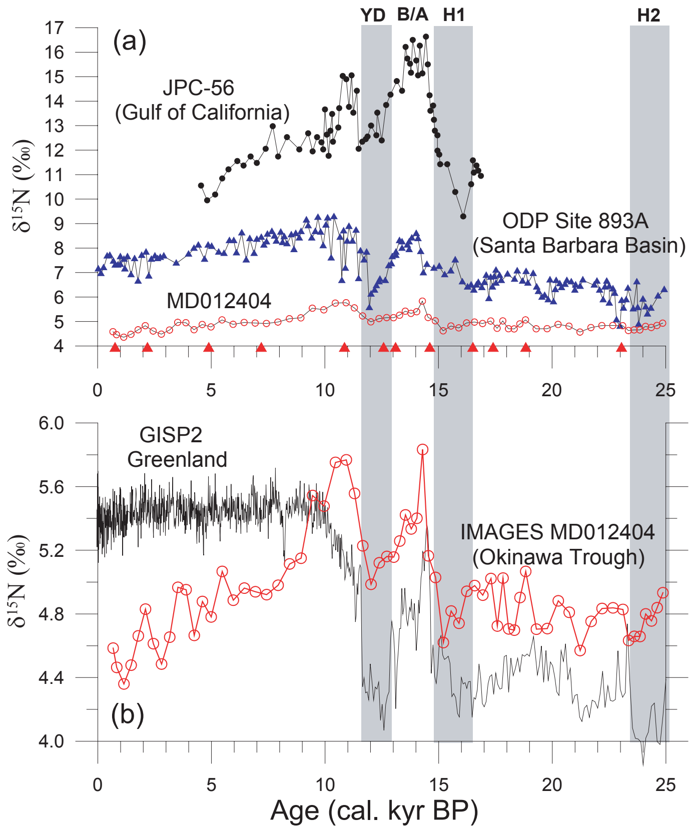

Fig. 3. (a) Temporal trends of sedimentary δ^{15}N for MD012404 (○), JPC-56 in the Gulf of California (•) and ODP 893A in the Santa Barbara Basin (▲). **(b)** Same as (a) but with different Y-axis for comparison. Record of δ^{18}O for GISP2 ice core (black curves) is also shown in (b). Bøling-Allerød warming period (B/A) is marked.

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